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Estimating the Mass of Pu-239 Waste Near P9-20 Probe Hole for the OU 7-10 Glovebox Excavator Method Project

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ABSTRACT

A review was conducted during the first week of September 2002 of previous passive gamma probe hole data analyses concerning the Pu-239 mass located near the P9-20 probe hole in the Subsurface Disposal Area. The review considered several analyses performed by P. Kuan, Y. Harker, W. Yoon, and N. Josten. Unfortunately, the logging data do not uniquely define Pu-239 waste mass or its distribution. The previous analyses constrained the Pu-239 mass values between 100 g and 2,520 g. Because of the wide range of values and their impact on the safe excavation of the area, *this review was conducted to determine the range of values for a single distributed source of Pu-239 mass*. Based on this review and key assumptions, it was determined that a lower value of 319 g for plutonium is consistent with the probe hole logging data. An upper value of 2,217 g is also consistent with the data, and could be considered as a reasonable upper bound on the mass. It must be noted that the conclusions of this review depend highly upon the initial assumptions made in the data analyses, and that the mass values reported in this review are based solely upon the logging data and the assumed accuracy of the model formulation.

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Estimating the Mass of Pu-239 Waste Near P9-20 Probe Hole for the OU 7-10 Glovebox Excavator Method Project

1. INTRODUCTION

A committee was convened in September 2002 to review the results of several separate analyses of nuclear spectroscopic logging data obtained in Pit 9 at the Subsurface Disposal Area within the Radioactive Waste Management Complex of the Idaho National Engineering and Environmental Laboratory. The objective of the review was to determine the range of worst-case mass values for a single distributed source of plutonium buried in the vicinity of borehole P9-20 in support of the Operable Unit (OU) 7-10 Glovebox Excavator Method Project. The committee checked that the different analyses were consistent, reasonable, and if estimates of maximum plutonium material could be determined from the logging data. This report is the result of reviewing the analyses of several scientists at the INEEL (P. Kuan, W. Yoon, and Y. Harker) and that of an external consultant N. Josten. This report represents the committee best evaluation of their analysis.

2. CHRONOLOGY

In January 2000, passive gamma logging measurements were conducted in the P9-20 probe hole. The original gamma spectra and analysis by Waste Management Technical Services (WMTS) were presented in EDF-ER-207, Appendix A, "Geophysical Logging at Pit 9, INEEL" (Josten 2000). The original analysis indicated a greater than 100,000 nCi/g concentration of Pu-239 at roughly a 6-ft depth. The original analysis was based on logging probe calibrations assuming that the radiation-producing source, or waste volume, was an infinitely large, homogeneously distributed matrix centered about the single probe hole. A subsequent analysis by Kuan (Attachment A) allowed for a single spherical distributed source located at some distance from the probe hole. This analysis shows large radiation source masses were consistent with the data. It is obvious that the allowance for attenuating media between the source and detector would increase the upper Pu-239 mass. Also at this time, Harker performed an analysis with an independent model of the Pu-239 mass (Appendix C in Attachment A) that is in basic agreement with the Kuan analysis. During May and June of 2000, additional probe holes were installed and logged in an area surrounding P9-20 to bound the mass determination. Two new analyses incorporating the data from the additional probe holes have been performed. Yoon's inverse MCNP analysis indicates a significantly reduced Pu-239 mass (Attachment B), while Josten's analysis is in basic agreement with the earlier Kuan and Harker models (Attachment C).

3. MODELS

All of the analyses considered in this review, with the exception of the Harker analysis for which there are few details available, appear to be sound in their methodology. They all make reasonable assumptions and seem accurate in their execution. However, the analyses rely on a set of assumptions about the Pu-239 particle size and soil density. These assumptions are by far the most significant assumptions in determining the overall mass. With the exception of the Yoon analysis, the analyses are a conservative estimate of the total mass contained. It must be mentioned that none of the reviewed models and analyses can give a unique solution. This means a wide range of Pu-239 mass is consistent with the logging data. The present review incorporates a χ^2 fitting criteria to the data analyses in an attempt to quantify the degree to which the model predicts the observed gamma emissions. Note, due to time limitations, no attempt was made to rigorously exhaust a complete range of the free parameters. A cursory parameter variance was performed to understand the models and place bounds on the mass. It is entirely possible, however, that other locations, masses, and sizes of the waste volume could fit the observed data equally well.

3.1 Kuan Analysis

The Kuan analysis (Attachment A) is based on data from a single probe hole, P9-20. There is a significant limitation in the original WMTS analysis. It assumes an infinite, homogeneously distributed radioactive source centered about the probe hole. The limitation exists because the calibration of the logging tool is reduced to a single conversion factor in nCi/g/c/s. Any accuracy of that calibration depends solely upon how closely the unknown geometry and waste matrix matches that of the logging tool calibration standard. Kuan's model essentially converts the original calibration to permit a discrete source removed from the probe hole by calculating the overlap of the waste volume and detector surface area elements. Kuan's model also allows for the gamma intensity attenuation from a soil layer of assumed density between the source and detector. Kuan's analysis concludes that an arbitrarily sized source could lead to an arbitrarily large mass of Pu-239. The choice was made to assume a waste volume matching that of a 55-gal drum (22.85 cm in diameter). With that assumption, Kuan concludes that a 2,000-g source of Pu-239 is consistent with the single probe hole data (based on the January 2000 logging data). Kuan's analysis also concluded that a 100-g source is consistent with the same single probe hole data (based on the January 2000 logging data).

3.2 Harker Analysis

The Harker analysis (Appendix C in Attachment A) was also based on the January 2000 logging data from the single probe hole log at P9-20. Harker used the three-dimensional code REMSPAT to solve for an arbitrary source volume that would account for the total counts observed with the passive gamma tool. By assuming the same waste volume as Kuan, he concluded that a 2,520-g source is possible. He also concluded that a second source slightly deeper in the burial site was needed to accurately fit the logging data. Extremely limited information about the Harker analysis was available for this review. There are some serious concerns expressed in Harker's report about the validity of the dead time corrections used in the original January 2000 logging data set. These concerns are supported by a revised analysis by WMTS that acknowledge the dead time errors. It is unclear to what extent the actual measured mass values can be trusted in this analysis and the original Kuan analysis.

3.3 Yoon Analysis

Yoon's analysis was the inverse solution of a Monte Carlo N-Particle transport model (Attachment B). It is the first analysis of this area to include the data from the additional probe holes surrounding P9-20 and the later May 2001 logging data. The data from the additional probe holes and the new P9-20 data have improved dead-time corrections. Even a casual look at the surrounding probe hole data indicates that the major radioactive source is not centered about the P9-20 hole, but is certainly closer to it than it is to the other probe holes. The surrounding probe hole data effectively eliminate the possibility that Kuan considered of an arbitrarily large source located an arbitrarily large distance away from P9-20. Yoon's work shows that the data are compatible with multiple low-mass sources adjacent to the probe. It appears from examining the results and from personal communication with Yoon, that this analysis is incomplete. A single iteration of two different source distributions was considered. It would be desirable to complete this analysis by varying the source distribution and by modifying the fit criteria to treat all of the probe holes with equal weight. It would also be desirable to use a source distribution matching that of the Josten analysis (Attachment C) to compare the results. Yoon's analysis could well prove to be the most accurate model when completed. It is a much more rigorous approach than the "back of the envelope" methods used by Kuan and Josten.

3.4 Josten Analysis

The Josten analysis used the new May 2001 logging data from P9-20 and additional surrounding probe holes (Attachment C). The principal benefit of the new data and additional probe log data is that they bound the amount of plutonium that could fit the logging data. Josten has accurately taken the basic formalism of the Kuan model and applied it to fit two additional adjacent probe holes logs simultaneously with P9-20 logged data. With the bounding data of the additional probe holes, Josten's 3-well analysis can also place an upper bound on the waste size and radioactive activity. Josten's analysis assumes a worst-case-highest Pu-239 mass by using a single spherical waste volume located off-center of P9-20. The probe hole logging data support this geometry with an indication of greatly reduced 414-keV gamma emissions from the surrounding probe holes. This review focused on Josten's analysis approach.

4. DISCUSSION

All of the analyses appear well thought out and scientifically sound. They all suffer from non-unique solutions. Any number of positions, source mass, size, or soil density combinations of the waste can yield an equally good fit to the data for a given set of assumptions. This review has attempted to quantify the data fit by introducing a χ^2 criteria with equal weighting to all of the probe holes. The parameter variation has, though, been only a cursory one. It is possible that a set of parameters could be found that yield a higher mass than those reported. However, by varying these parameters, the relationship between the parameters and how each individual variance affects the mass estimate could be explored.

4.1 Worst-Case Scenario

The Josten analysis assumes a single, spherical homogenous source at a depth of 6 ft between the three probes P9-20, P9-20-01, and P9-20-06. The logging data strongly support this interpretation. However, while examining the effect of varying the parameters in the Josten analysis, it appears that the inclusion of additional sources would yield a better data fit. This was not performed. The inclusion of additional radioactive sources nearer the P9-20-01 and P9-20-06 probes could greatly reduce the Pu-239 mass estimate. By not including additional sources, the Josten analysis calculates the highest mass estimate possible, creating a worst-case scenario. It is obvious that additional sources would decrease the mass estimate. The best χ^2 fit to the logging data gives an estimate of 1,689 g of Pu-239 contained within a 10-cm-radius sphere whose edge is approximately 10.5 cm away from P9-20 borehole (Fig 1). An equally good fit is obtained by assuming a slightly lower soil density resulting in a nominal 1,081 g-source of Pu-239 located 9.5 cm away from P9-20 (Fig. 2). Likewise, a reasonable fit can also be obtained with a much reduced soil density and only 243 g of Pu-239 placed 5 cm from P9-20 (Fig. 3). All of these solutions will fit the data. These results, self-shielding effects, and soil density dependence discussed below, indicate the importance of the initial assumptions.

4.2 Initial Assumptions

Two very reasonable but basic assumptions are made in all of the analyses that greatly affect the final Pu-239 mass estimate. The first is the particle size of any chunk of Pu-239 found in the waste. Because of the high density and absorption in Pu, a very significant amount of self-shielding occurs if the particle size exceeds 1,000-μm spherical diameter, about the size of a small BB. At 1,000 μm, the gamma count rate will underestimate the mass by 20%, meaning the observed mass activity is 80% actual. At 1 cm diameter, the mass can be underestimated by a factor of 4, and at 5 cm diameter, the mass is underestimated by a factor of 20. The waste processing and shipping records strongly indicate that the Pu content is composed of small pieces or thin films where self-shielding would be negligible. However, the reviewers find it prudent to allow for a 1,000-µm particle size and thereby *increase the mass estimates by a factor of 25%* to account for 20% self-shielding effect (i.e. the self-shielding correction is 1/0.8 = 1.25). For these calculations, the Pu particles were assumed to be uniformly distributed in a waste sphere matrix of bulk density 2 g/cc, consistent with Pu embedded in a graphite mold. The buried waste inventory and logging data indicate a graphite waste form may be present.

The second basic assumption that greatly affects the mass estimate is the soil density. A direct sampling[a] indicates a mean bulk density 1.44 g/cm^3 for disturbed soils at SDA (Figure 4). This is consistent for sandy soil with 46% porosity. This is a surprising lack of compaction given the time that the P9-20 area soil has had to subside. However, video images from a visual probe next to P9-20 indicate that a substantial amount of void space could be present in the waste layer. It is difficult to quantify the density in the waste layer by the visual probes, but it is reasonable to assume that the gross soil density could be reduced by the introduction of substantial void space. A conservative estimate of half-liquid saturation, which is consistent with soil moisture estimates of 20 to 26% in the waste zone, raises the downhole soil density estimate to 1.64 g/cm^3. Differential-attenuation of multiple gamma ray emissions from Pu-239 analysis was performed by Kuan, indicating a slightly lower average soil density of 1.53 g/cm^3. However, the details of that calculation are unknown to this review.

4.3 Data Quality

Since the probe holes were originally calibrated in an assumed geometry of uniform waste distribution, the logging data were reported in terms of concentration nCi/c/s for an infinite homogeneous Pu-239 distribution. The adaptation of the models allowing for different geometries requires the reported concentration to be converted back to counts per second. As a check on the calibration-conversion, this review examined original logging spectral data and fit the Pu-239 414-keV characteristic gamma ray peak from the 6-ft depth level. In each case checked, the observed count rate reported in the Josten analysis is lower by 5% than the original spectra indicated. It is unknown whether an additional calibration factor was included to extract the Josten values. It is deemed conservative to adopt an *additional* 5% mass increase to the reported upper limits to account for count rate corrections (i.e., the count rate correction is 1/0.95 = 1.05).

The combined self-shielding and count rate corrections (1.25 x 1.05)= 1.31 will be applied in the next section to the estimated mass results from the Josten 3-well implementation of Kuan's model.

4.4 Existing Records

This review makes no comment on the existing processing and shipping records, except to say that it has been reported that nondestructive assays on above-ground storage drums of the same waste stream have been found containing as much as 310 g of Pu-239 (Nielsen 2002). With this in mind, the upper self-absorption and count rate corrected mass limits of 1,689 g (x 1.25 x 1.05) = 2,217 g seems high. However, this value cannot be ruled out by the existing data. The nominal value 1,081 g (x 1.25 x 1.05) = 1,419 g also seems high, but also cannot be ruled out as a possibility. The 243g (x 1.25 x 1.05) = 319 g would be close to the highest mass found in any aboveground storage drum, and is also consistent with

a. Nick Josten, private communication.

the data. Due to the poor fit to multiple borehole data, a 104-gram source is deemed not consistent with the measured data even when a low soil density is assumed (Figure 5).

5. CONCLUSIONS

The best estimates of this limited review are based on applying Josten's analysis to the measured 414-keV passive gamma logging data with the limitations of the assumptions discussed above. The range of worst-case mass estimates for a single distributed source of Pu-239-waste has a lower estimate of 319 g located 5 cm from P9-20, in a fairly loosely compacted soil, to an upper estimate of 2,217 g located 10 cm from P9-20. These mass estimates are examples of "worst-case" scenarios since the single-sphere model preferentially places the Pu mass as far from the detectors as possible. It is believed that considerably lower mass estimates can be made if multiple sources of Pu or large void spaces are allowed in the analyses. As proposed by Harker and Yoon, these assumptions can be tested with a full 3D model.

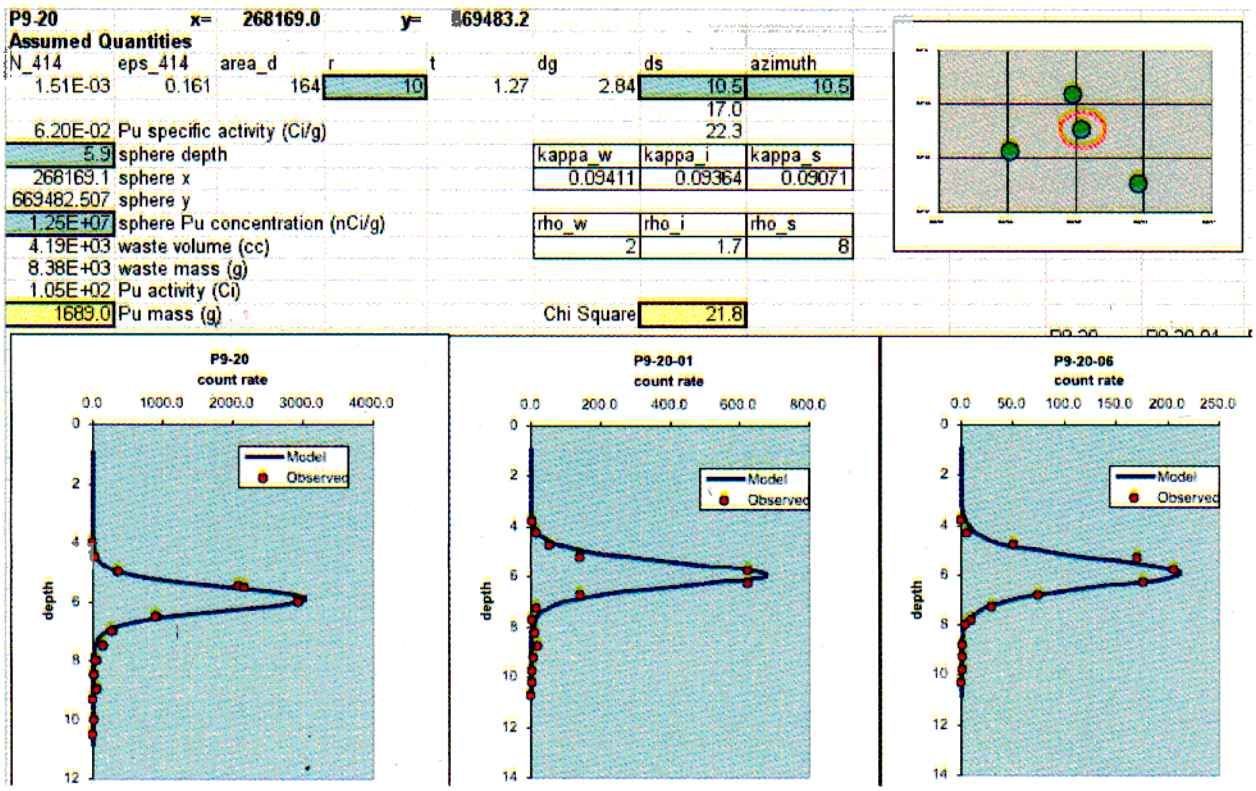

Figure 1. Josten analysis and logging data fits for the P9-20, P9-20-01, and P9-20-06 probe holes passive gamma data. A Pu-239 mass of 1,689 g fits the data with $\chi^2 = 21.8$ with the initial assumption of 1.7-g/cm^3 soil density.

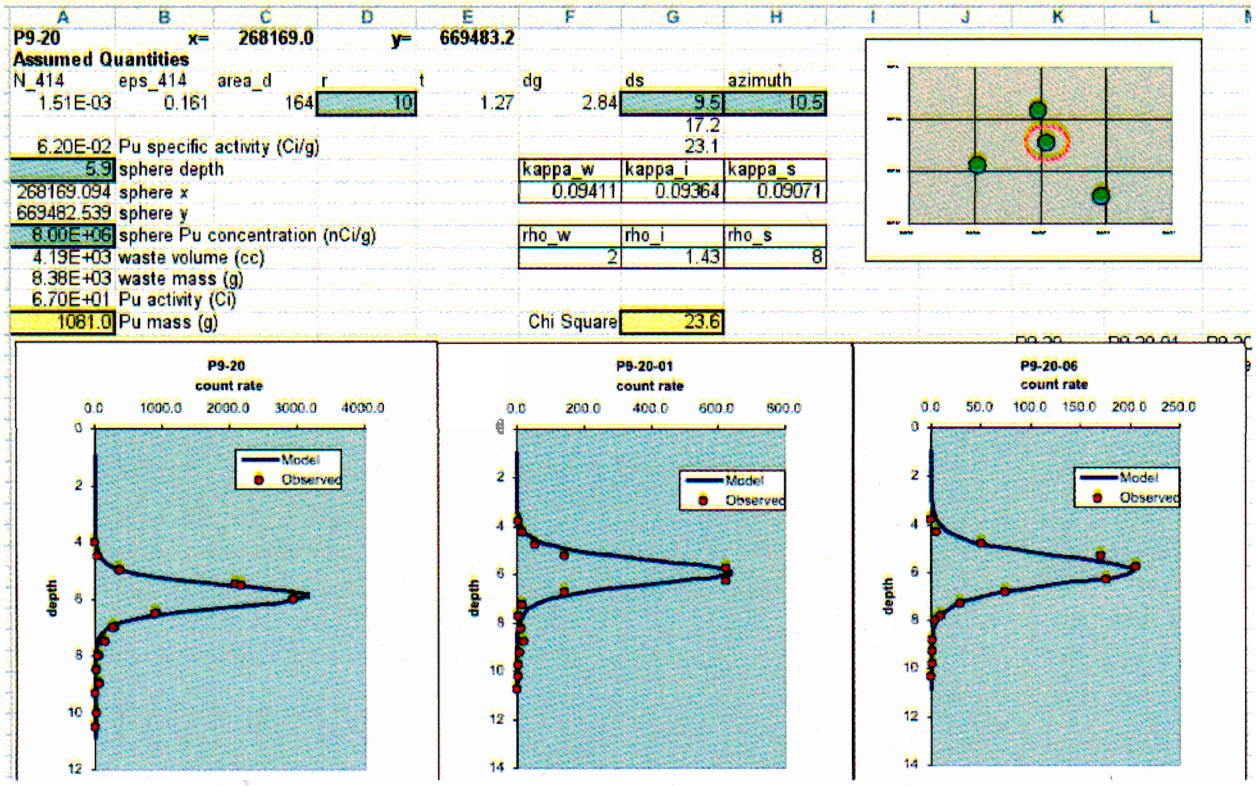

Figure 2. Josten analysis and logging data fits for the P9-20, P9-20-01, and P9-20-06 probe holes passive gamma data. A Pu-239 mass of 1,081 g fits the data with $\chi^2 = 23.6$ with the initial assumption of 1.43-g/cm^3 soil density.

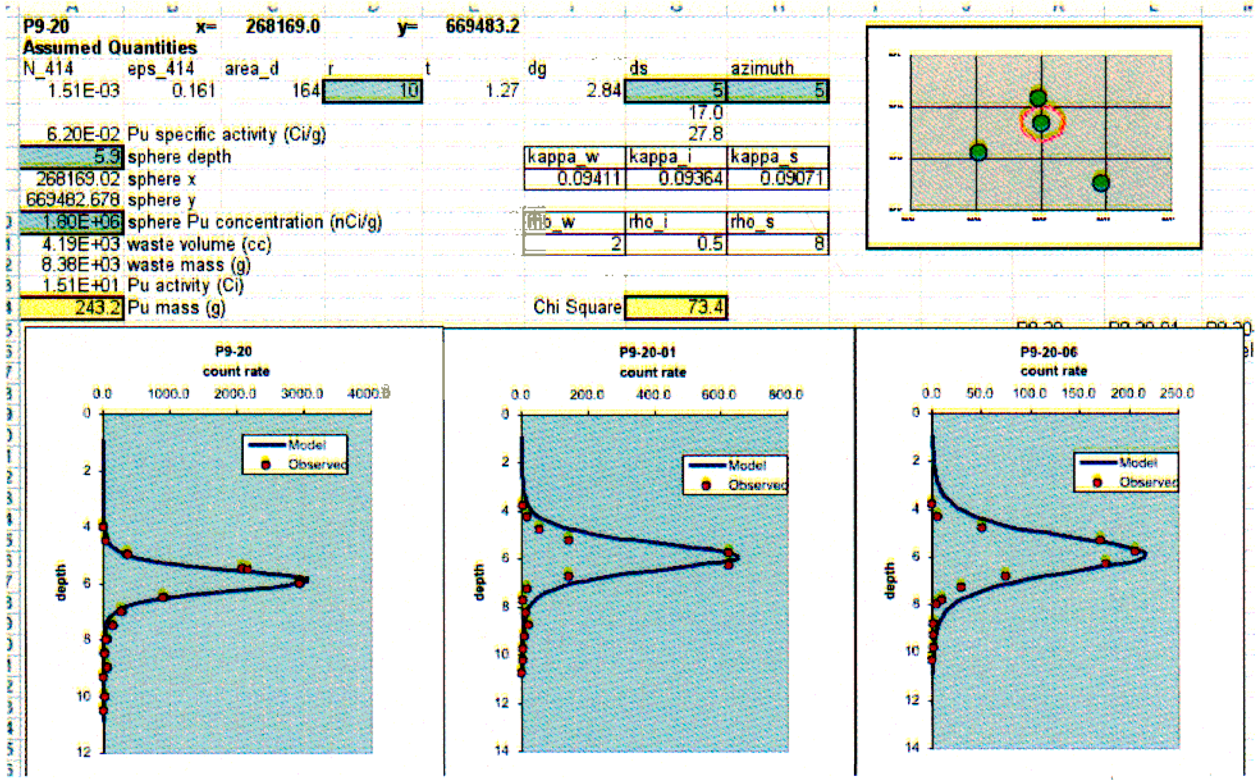

Figure 3. Josten analysis and logging data fits for the P9-20, P9-20-01, and P9-20-06 probe holes passive gamma data. A Pu-239 mass of 243 g fits the data with $\chi^2 = 73.4$ with the initial assumption of 0.5-g/cm^3 soil density.

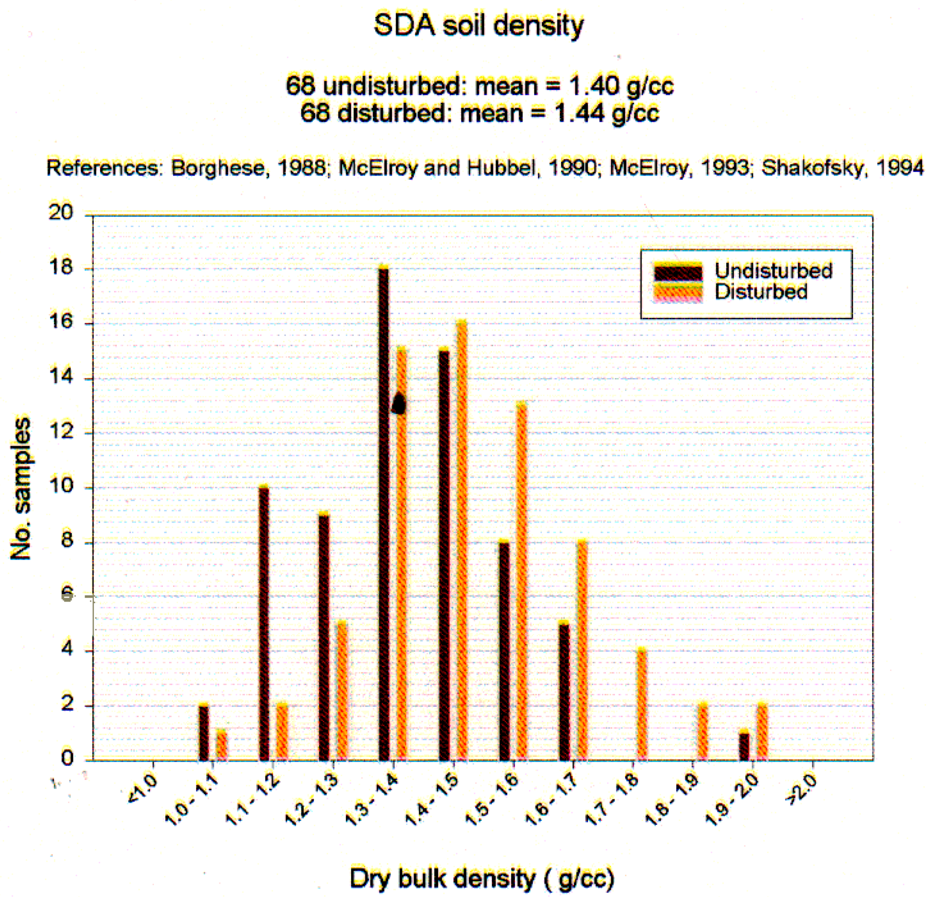

Figure 4. Summary of soil densities from past sampling activities at the SDA.

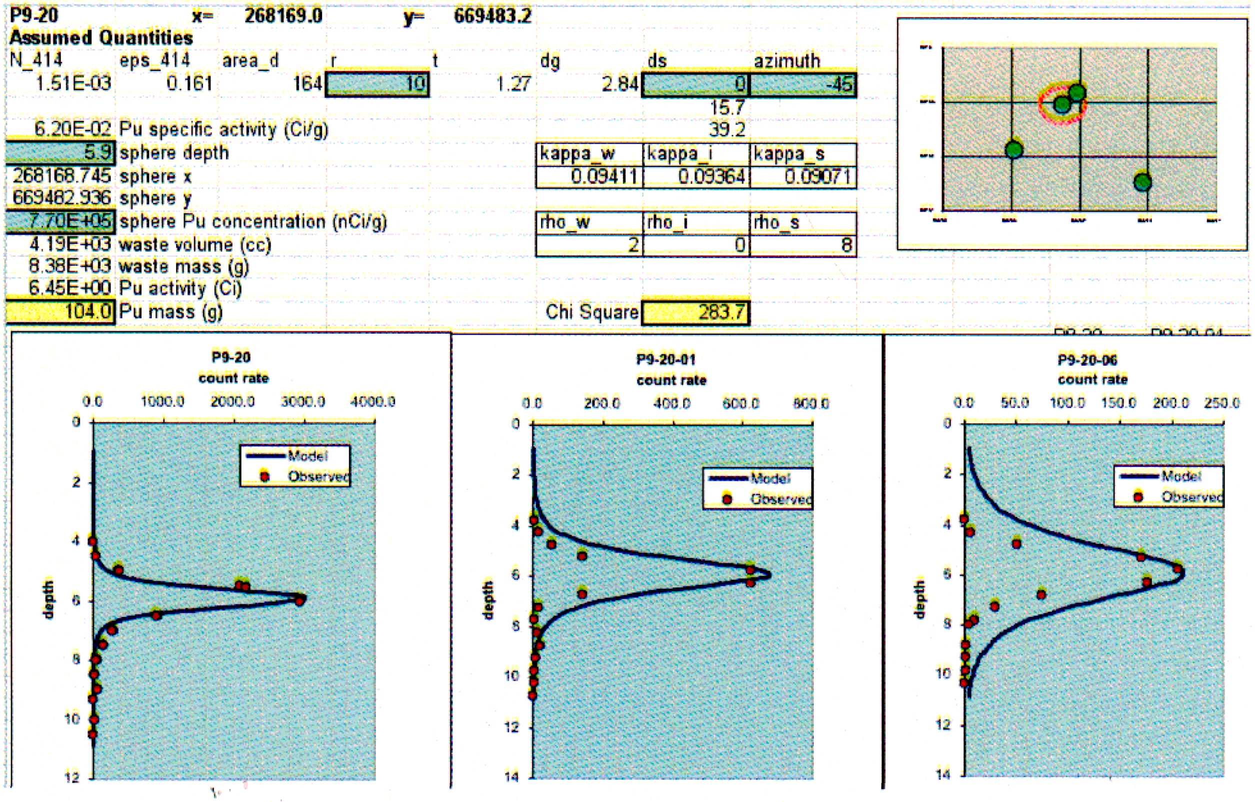

Figure 5. Josten analysis and logging data fits for the P9-20, P9-20-01, and P9-20-06 probe holes passive gamma data. A Pu-239 mass of 104 g cannot fit the data even with the initial assumption of 0.0-g/cm^3 soil density.

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